PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference LU6073/CB	FOR FURTHER ACTION	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
International application No. PCT/EP 03/13553	International filing date *(day/month/year)* 02.12.2003	Priority date *(day/month/year)* 06.12.2002
International Patent Classification (IPC) or both national classification and IPC C08F4/64		
Applicant BASELL POLYOLEFINE GMBH et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 7 sheets, including this cover sheet.

   ☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

   These annexes consist of a total of 5 sheets.

3. This report contains indications relating to the following items:

   - I ☒ Basis of the opinion
   - II ☐ Priority
   - III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   - IV ☒ Lack of unity of invention
   - V ☒ Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
   - VI ☐ Certain documents cited
   - VII ☐ Certain defects in the international application
   - VIII ☐ Certain observations on the international application

Date of submission of the demand	Date of completion of this report
28.06.2004	30.03.2005
Name and mailing address of the international preliminary examining authority: European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized Officer Kirsch, C Telephone No. +49 89 2399-2191

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/EP 03/13553

I. Basis of the report

1. With regard to the **elements** of the international application *(Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17))*:

Description, Pages

1-31	as originally filed

Claims, Numbers

3-9	as originally filed
1-2	filed with telefax on 19.11.2004

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

   These elements were available or furnished to this Authority in the following language: , which is:

   ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).

   ☐ the language of publication of the international application (under Rule 48.3(b)).

   ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

   ☐ contained in the international application in written form.

   ☐ filed together with the international application in computer readable form.

   ☐ furnished subsequently to this Authority in written form.

   ☐ furnished subsequently to this Authority in computer readable form.

   ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

   ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

   ☐ the description, pages:

   ☐ the claims, Nos.:

   ☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)).

   (Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

IV. Lack of unity of invention

1. In response to the invitation to restrict or pay additional fees, the applicant has:

   ☐ restricted the claims.

   ☐ paid additional fees.

   ☐ paid additional fees under protest.

   ☐ neither restricted nor paid additional fees.

2. ☐ This Authority found that the requirement of unity of invention is not complied with and chose, according to Rule 68.1, not to invite the applicant to restrict or pay additional fees.

3. This Authority considers that the requirement of unity of invention in accordance with Rules 13.1, 13.2 and 13.3 is

   ☒ complied with.

   ☐ not complied with for the following reasons:

4. Consequently, the following parts of the international application were the subject of international preliminary examination in establishing this report:

   ☒ all parts.

   ☐ the parts relating to claims Nos. .

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	1-9
	No:	Claims	
Inventive step (IS)	Yes:	Claims	
	No:	Claims	1-9
Industrial applicability (IA)	Yes:	Claims	1-9
	No:	Claims	

2. Citations and explanations

   see separate sheet

Reference is made to the following documents :

D1: EP-A-0 743 317
D2: WO02/18397
D3: WO98/40419
D4: WO99/40129
D5: EP-A-0 576 970

The subject-matter of the present application deals with biscyclopentadienyl transition metal compounds for use as catalysts in the polymerization of olefins.

Re Item I
Basis of the report

The amendments filed with telefax of 19.11.2004 limit the subject-matter of the claims to a specific sub-class of compounds of formula (I), namely to bis(4-arylindenyl) metal derivatives. Thus, one originally disclosed meaning was deleted from a list of sizeable length specifying possible alternative meanings of R2 within a generic chemical formula defining in its turn a claimed class of chemical compounds. Whereas any limitation necessarily implies that what remains is less than what was available before the limitation, the present deletions did not result in singling out a particular combination of specific meanings, i.e. any hitherto not specifically mentioned individual compound or group of compounds, but maintained the remaining subject-matter as a generic group of compounds differing from the original group only by its smaller size, the number of encompassed compounds having been indeed reduced as a consequence of the said deletions. In the present situation, this shrinking of the generic group of chemical compounds is not objectionable under Article 34(2)b PCT, since these deletions did not lead to a particular combination of specific meanings of the respective residues which was not disclosed originally. Furthermore, the amended claim 1 is supported by the examples since all the originally filed examples relating to a compound of the invention are still within the scope of the amended claim 1.

Re Item IV
Unity of invention

The compounds of formula (I) possess a biscyclopentadienyl moiety at least disubstituted in 4- and 7-positions. The subject-matter has been specifically limited to bis(4-arylindenyl) metal derivatives, said compounds being useful as catalysts in the polymerization of olefins. The common structural element is now represented by bis(4-arylindenyl) metal core structure with a further substituent in 7-position. Since document D1 only reveals 4-heteroarylindene derivatives, the structural element is novel, and with the polymerization catalytic activity, represent the single unitary inventive concept linking together the different subject-matter. The newly filed set of claims can therefore be regarded as unitary.

Re Item V

Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Document D1 discloses biscyclopentadienyl transition metal compounds of formula (II) useful catalysts for the polymerization of olefins. Formula (II) of D1 partially overlaps with the present claimed formula (I) when R^{10} of D1 is an aryl moiety, R5 is (CR^8R^9-CR^8R^9), n is 1 and R^{13} is not hydrogen. This combination of these different features is not expressively disclosed in D1 and represent therefore a new structural element common to all the alternative comprised in the overlap which was not disclosed in the prior art. The subject-matter of present claims 1-9 is considered as a new selection with regard to document D1 (Art. 33(2) PCT).

   Document D2 describes biscyclopentadienyl transition metal compounds of formula (II) useful catalysts for the polymerization of olefins. General formula (I) of D2 overlaps with formula (I) of claim 1 of the present application when, according to D2, R^3 represents an aryl, R^6 is not H and I=j=1. This specific combination of features (aryl group in 4-position, the presence of a further substituent in 7-position and ethylidene linker between the two biscyclopentadienyl moieties) is not explicitly disclosed in D2 and represents a new technical element common to all the alternatives comprised in the overlap. Novelty can therefore be acknowledged with regard to document D2 (Art. 33(2) PCT).

   Document D3 reveals a process of synthesis of olefin polymers wherein a biscyclopentadienyl transition metal catalyst of formula (I) is used. There is also an overlap between the subject-matter of D3 and the present invention. However, the

specific combination aryl group in 4-position + substituent in 7-position + ethylidene linker between the two biscyclopentadienyl moieties is not explicitly described in D3. This specific structural element provides a contribution over the prior art document D3. Novelty is also established vis-à-vis D3 (Art. 33(2) PCT).

Document D4 deals with a catalyst system containing, amongst others, a metallocene compound of formula (VI). Formula (VI) of D4 partially overlaps with the present claimed subject-matter. Examples with an ethylidene linker and an aromatic ring in 4-position are provided (see p. 15, l. 2-14 for instance). However, such structural features with the presence of a further substituent in 7-position is not explicitly revealed in D4. The combination of these features therefore provides a contribution over the prior art D4. Novelty is acknowledged with regard to document D4 (Art. 33(2) PCT).

Document D5 discloses biscyclopentadienyl metallocene catalysts with an aromatic group in 4-position. Formula (I) of D5 overlaps with the present invention when R^{13} is an ethylidene linker and R^7 is not hydrogen. The presence of an aromatic ring in 4-position is disclosed in example G. However, the combination of this specific structural element with a further substituent in 7-position is not described in D5. This combination represent a new technical teaching which was disclosed in the prior art document D5. Novelty is also established with regard to document D5 (Art. 33(2) PCT).

The subject-matter of present claims 1-9 represent a new selection vis-à-vis de technical teaching revealed in D1 to D5 (Art. 33(2) PCT).

2. Document D1, which is considered to represent the most relevant state of the art, discloses biscyclopentadienyl transition metal compounds of formula (II) and their use as catalysts for the polymerization of olefins. D1 also reveals the combination of an aromatic substituent in 4-position with the presence of a further substituent in 7-position. The present invention represent a specific combination of structural feature generally disclosed in document D1.

The problem to be solved by the present application may be considered as the provision of further biscyclopentadienyl metallocene derivatives useful as polymerization catalysts.

The subject-matter of claims 1-9 consists in the selection of a particular class of compounds from the general formula disclosed in D1. Such a selection can only be regarded as inventive, if the claimed compounds presents unexpected effects or properties in comparison with the compounds of the prior art. However, no such effects or properties are indicated in the application. Hence, no inventive step is present in the subject-matter of claims 1-9.

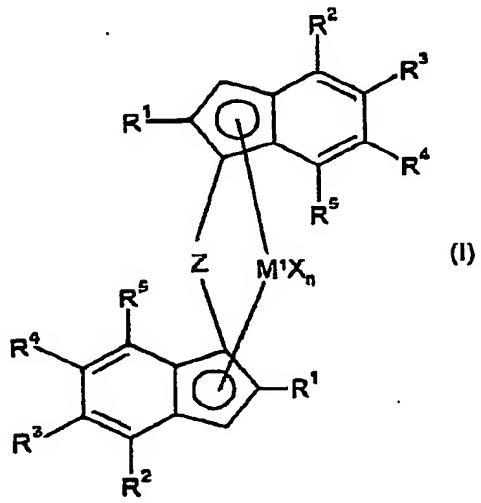
3. Contrary to the requirements of Rule 5.1(a)(ii) PCT, the relevant background art disclosed in document D1 is not mentioned in the description, nor is this document identified therein.

The newly filed claim 2 has not been amended accordingly to pending claim 1 and refers to definitions which are not anymore encompass by the scope of claim 1 (Art. 6 PCT).

new claim 1

Claims:

1. An organometallic transition metal compound of the formula (I)



(I)

where

M^1 is an element of group 3, 4, 5 or 6 of the Periodic Table of the Elements or the lanthanides,

X are identical or different and are each halogen, hydrogen, C_1-C_{20}-alkyl, C_2-C_{20}-alkenyl, C_6-C_{22}-aryl, alkylaryl or arylalkyl each having from 1 to 10 carbon atoms in the alkyl part and from 6 to 22 carbon atoms in the aryl part, $-OR^6$ or $-NR^6R^7$, where two radicals X may also be joined to one another,

n is a natural number from 1 to 4 which corresponds to the oxidation number of M^1 minus 2,

R^1 is a cyclic, branched or unbranched C_1-C_{20}-alkyl radical, a C_2-C_{20}-alkenyl radical, an arylalkyl radical having from 1 to 10 carbon atoms in the alkyl part and from 6 to 22 carbon atoms in the aryl part or a C_4-C_{24} heteroaromatic radical selected from the group consisting of substituted or unsubstituted thienyl radicals or of substituted or unsubstituted furyl radicals,

R^2 is a substituted or unsubstituted C_6-C_{40}-aryl radical.

R^3 is hydrogen or a cyclic, branched or unbranched C_1-C_{20}-alkyl radical, a C_2-C_{20}-alkenyl radical, an arylalkyl radical having from 1 to 10 carbon atoms in the alkyl part and from 6 to 22 carbon atoms in the aryl part,

R^4 is hydrogen or a cyclic, branched or unbranched C_1-C_{20}-alkyl radical, a C_2-C_{20}-alkenyl radical, an arylalkyl radical having from 1 to 10 carbon atoms in the alkyl part and from 6 to 22 carbon atoms in the aryl part,

R^5 is a cyclic, branched or unbranched C_1-C_{20}-alkyl radical, a C_2-C_{20}-alkenyl radical, an arylalkyl radical having from 1 to 10 carbon atoms in the alkyl part and from 6 to 22 carbon atoms in the aryl part,

and

Z is a divalent group CR^8R^9-$CR^{10}R^{11}$, where R^8, R^9, R^{10} and R^{11} are identical or different and are each hydrogen, a trimethylsilyl group, a C_1-C_{10}-alkyl group, a C_1-C_{10}-fluoroalkyl group, a C_6-C_{10}-fluoroaryl group, a C_6-C_{10}-aryl group, a C_8-C_{40}-arylalkenyl group, a C_7-C_{40}-arylalkyl group or a C_7-C_{40}-alkylaryl group or two adjacent radicals together with the atoms connecting them may also form a saturated or unsaturated ring having from 4 to 15 carbon atoms.

2. An organometallic transition metal compound of the formula (I) as claimed in claim 1,

where

M^1 is an element of group 4 of the Periodic Table of the Elements,

n is 2,

R^1 is C_1-C_{10}-alkyl,

R^3 is hydrogen or a C_1-C_{10}-alkyl radical,

R^4 is hydrogen or a C_1-C_{10}-alkyl radical,

R^5 is a C_1-C_{10}-alkyl radical and

Z is CH_2-CH_2.

32a

A)

~~The radical R^1 is hydrogen or a C_1-C_{40} radical.~~ R^1 is ~~preferably~~ a cyclic, branched or unbranched C_1-C_{20}-~~[preferably C_1-C_8-]~~alkyl radical, a C_2-C_{20}-~~[preferably C_2-C_8-]~~alkenyl radical, an arylalkyl radical having from 1 to 10~~[preferably from 1 to 4]~~ carbon atoms in the alkyl part and from 6 to 22~~[preferably from 6 to 10]~~ carbon atoms in the aryl part or a ~~C_2-C_{40}, preferably C_3-C_{20}~~ heteroaromatic radical~~[, particularly]~~ selected from the group consisting of substituted or unsubstituted thienyl radicals or of substituted or unsubstituted furyl radicals~~[. Examples of particularly preferred radicals]~~

B

~~The radical R^3 is hydrogen or a C_1-C_{40} radical.~~ R^3 is ~~[preferably]~~ hydrogen or a cyclic, branched or unbranched C_1-C_{20}-~~[preferably C_1-C_{10}-]~~alkyl radical, a C_2-C_{20}-~~[preferably C_2-C_8-]~~alkenyl radical, an arylalkyl radical having from 1 to 10~~[preferably from 1 to 4]~~ carbon atoms in the alkyl part and from 6 to 22~~[preferably from 6 to 10]~~ carbon atoms in the aryl part~~[. Examples of particularly pre-]~~

R^4 ~~is hydrogen or a C_1-C_{40} radical,~~ page 33 a C)

R^5 ~~is a C_1-C_{40} radical~~, page 33 a D)

and

Z is a divalent group CR^8R^9-$CR^{10}R^{11}$, where R^8, R^9, R^{10} and R^{11} are identical or different and are each ~~hydrogen or a C_1-C_{40} radical~~ page 33a E)

2. An organometallic transition metal compound of the formula (I) as claimed in claim 1,

where

M^1 is an element of group 4 of the Periodic Table of the Elements,

n is 2,

R^1 is C_1-C_{10}-alkyl,

R^3 is hydrogen or a C_1-C_{10}-alkyl radical,

R^4 is hydrogen or a C_1-C_{10}-alkyl radical,

R^5 is a C_1-C_{10}-alkyl radical and

Z is CH_2-CH_2.

C)

~~The radical R^4 is hydrogen or a C_1-C_{40} radical. R^4~~ is ~~preferably~~ hydrogen or a cyclic, branched or unbranched C_1-C_{20}-~~[preferably C_1-C_{10}-]~~alkyl radical, a C_2-C_{20}-~~[preferably C_2-C_8-]~~alkenyl radical, an arylalkyl radical having from 1 to 10~~[preferably from 1 to 4]~~ carbon atoms in the alkyl part and from 6 to 22~~[preferably from 6 to 10]~~ carbon atoms in the aryl part, ~~Examples of particularly pre-~~

D)

~~The radical R^5 is a C_1-C_{40} radical. R^5~~ is ~~preferably~~ a cyclic, branched or unbranched~~[preferably unbranched,]~~ C_1-C_{20}-~~[preferably C_1-C_{10}-]~~alkyl radical, a C_2-C_{20}-~~[preferably C_2-C_8-]~~alkenyl radical, an arylalkyl radical having from 1 to 10~~[preferably from 1 to 4]~~ carbon atoms in the alkyl part and from 6 to 22~~[preferably from 6 to 10]~~ carbon atoms in the aryl part, ~~Examples of particularly pre-~~

E)

~~Z is a divalent group GR^8R^9-$GR^{10}R^{11}$, where R^8, R^9, R^{10} and R^{11} are identical or different and are [each hydrogen or a C_1-C_{40} radical. R^8, R^9, R^{10} and R^{11} are preferably]~~ each hydrogen, a trimethylsilyl group, a C_1-C_{10}-~~[preferably C_1-C_3-]~~alkyl group, a C_1-C_{10}-fluoroalkyl group, a C_6-C_{10}-fluoroaryl group, a C_6-C_{10}-aryl group, a C_8-C_{40}-arylalkenyl group, a C_7-C_{40}-arylalkyl group or a C_7-C_{40}-alkylaryl group, ~~or~~ two adjacent radicals together with the atoms connecting them may also form a saturated or unsaturated ring having from 4 to 15 carbon atoms.